Magnetic and thermodynamic properties of $Sr_2LaFe_3O_9$

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We use a Dirac-Heisenberg Hamiltonian with biquadratic exchange interactions to describe the first-order magnetic transition occurring in the perovskite $Sr_2LaFe_3O_9$. Up on fitting the experimental curve for the magnetic susceptibility below and above the Néel temperature, we give an estimate of the exchange integrals for the antiferromagntic and ferromagnetic interactions in this compound. Within linear spin-wave theory we find that the magnon spectrum comprises a gapless antiferromagnetic mode together with two gapped ferromagnetic ones.

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I. INTRODUCTION

Since the discovery of high-T_c copper-oxide superconductors [1], investigations of superconductivity and magnetism of d-electron based oxides with perovskite structure have gained new interest. Iron based oxides are not superconducting, but exhibit interesting electronic properties such as disproportionation as in $CaFeO_3$ or metalic properties as in $SrFeO_3$. In addition, these ferrites often shown very peculiar magnetic properties.

Almost stoichiometric $Sr_2LaFe_3O_{9-\delta}$, ($\delta \simeq 0$) can be prepared under air by long annealing at $200°C$ or using electrochemical oxidation in alkaline solution [2], [3], [4]. All reported data of structural and physical properties show very similar features.

The low-temperature Mossbauer data for the perovskite $Sr_2LaFe_3O_{8.94}$ studied in [2] are consistent with a 2 : 1 ratio of the two types of Fe cation with electronic characteristics fairly close to Fe^{3+} and Fe^{5+}, respectively. The Neutron-Powder-Diffraction data at $5K$, show clear evidence that the low temperature phase exhibits an antiferromagnetic ordering [2]. More precisely, there is antiferromagnetic ordering among subcells of ferromagnetically-ordered spins. At room temperature only is observed an average value of the charge, which demonstrates the existence of a fast electron transfer.

So, upon heating the compound passes from a mixed-valence phase through a first-order transition to the paramagnetic average-valence phase. Experimental data [3] for the specific heat show two peaks, the highest of which seems to be related to a first-order phase transition. The smaller peak, on the other hand, disappears as one fills in more oxygen vacancies in the sample, as was shown by the data of ref. [4] for the sample $Sr_2LaFe_3O_9$, where the specific heat only shows one (large) peak at about the same temperature as in [3].

The inverse magnetic susceptibility for $Sr_2LaFe_3O_{8.94}$ shows [2] an abrupt change at about $200K$ with a minimum indicative of a transition to antiferromagnetic ordering; and above this temperature the susceptibility is almost field-independent; while below there is a pronounced dependence on the magnetic field which appears to be an intrinsic property of the phase, and suggests the possibility of a (weak) ferromagnetism, probably due to spin mis-alignment. The same sharp jump in the magnetic susceptibility is reported in refs. [3] and [4].

According to previous structural studies [2], this compound exhibits an almost cubic perovskite structure, the rhombohedral distortion being very small and not detectable by X-Ray Diffraction analysis. The magnetic structure was determined from the Neutron-Diffraction data [2] and is shown in figure 1.

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The coupling between iron cations is of a superexchange-type via oxygen anions. However, the usual approximation consists in treating the system as that of iron cations interacting via an effective exchange integral. More precisely, in our case the coupling between two Fe^{3+} ions is antiferromagnetic with the exchange constant denoted henceforth by[1] J_a, while that between Fe^{3+} and Fe^{5+} is ferromagnetic with the constant J_f, while there is no coupling between two cations Fe^{5+}, as this would occur along the diagonal of the (slightly deformed) elementary cube, and thus would be negligible in comparison with the other two couplings (see figure 1). In our particular case, these exchange integrals are obtained by fitting the experimental data with a model of localized spins with two kinds of nearest-neighbor interactions.

Accordingly, we consider a model that describes a system of 6 localized spins, four of $\frac{5}{2}$ and two of $\frac{3}{2}$, based on a Dirac-Heisenberg Hamiltonian with antiferromagnetic and ferromagnetic couplings J_a, J_f, respectively and taking into account the corresponding biquadratic interactions, denoted by j_a, j_f. The latter are known [5] to induce a first-order transition because of the strong change of the exchange interactions as a function of the interatomic distances, and also the associated change in the elastic energy of the material. The characteristic and very significant feature of such magnetic-transformation mechanism is the sharp change of the elastic constants of the crystal in the absence of any volume changes in the region of the antiferromagnetic transformation [5]. In our case, the exchange-induced distortion of the crystal is of an order of magnitude too small [2], [3] to be responsible for the change of the order of the magnetic transition. In fact, it was pointed out in [6] and then shown in [7] that the usual superexchange mechanism is fully capable of explaining the origin and magnitude of the biquadratic exchange interaction. Also Nagaev ([8], section 2.7) studied the interplay between Dirac-Heisenberg and different kinds of non-Dirac-Heisenberg interactions and the role of the latter in changing the order of the magnetic transition viz in MgO, MnO and NiO. Moreover, since the spins in our case are much larger than $\frac{1}{2}$, more excited spin states start to participate in increasing the entropy of the system, and the two-level-system approximation no longer holds as the temperature increases. Therefore, in order to account for this effect, we must add higher-order terms to the Dirac–Heisenberg Hamiltonian. Accordingly, we shall add nearest-neighbor biquadratic interactions which, from the physical point of view could account for the effect of charge disproportionation occurring in our compound. Next, we show that indeed the biquadratic contributions to the exchange energy account for the first-order magnetic transition occurring in $Sr_2LaFe_3O_9$, and which is reflected by a sharp jump in the magnetic susceptibility and large peak in the specific heat. Consistently, we also predict an anomalous behavior for the sublattice magnetizations, i.e. the jump down to zero in the vicinity of the Néel temperature. Indeed, we obtain a good fit of the experimental data on the specific heat and the magnetic susceptibility in both the ordered and disordered phases for the exchange interactions $\frac{J_a}{k_B} \simeq 26K$, $\frac{J_f}{k_B} \simeq 6.5K$; and the corresponding biquadratic exchange integrals $j_a \simeq 0.11 \times J_a$ and $j_f \simeq 0.08 \times J_f$.

Note that the antiferromagnetic exchange integral J_a between two iron ions Fe^{3+} in perovskite compounds such as $LaFeO_3$ was also estimated by Anderson [10] who found $J_a \simeq 26K$, and a close value, $24K$, was also found by Grenier et al. [11] in $CaFe_2O_5$ with similar perovskite structure.

Next, we compute the magnon spectrum within the approach of linear spin-wave theory. The spectrum contains a gapless antiferromagnetic mode and two gapped ferromagnetic branches. Unfortunately, to the best of our knowledge, there are so far no experimental data on the Inelastic-Neutron Scattering for the present powder sample. However, to account for the anomalous behavior of the sublattice magnetizations near the transition point, it turns out that non-linear spin-wave corrections are necessary.

II. THEORY

a. Hamiltonian   The unit cell of our system is sketched in figure 1, and the Dirac-Heisenberg Hamiltonian for such system can be written as

$$H_{DH} = -J_f \sum_{\langle i,j \rangle} \sum_{\alpha,\beta} \mathbf{s}_{i\alpha} \cdot \mathbf{S}_{j\beta} + J_a \sum_{\langle i,j \rangle} \sum_{\alpha,\beta} \mathbf{S}_{i\alpha} \cdot \mathbf{S}_{j\beta} \qquad (1)$$

where henceforth $\mathbf{S}, \mathbf{s}$ denote the spins $\frac{5}{2}$ of Fe^{3+}, and $\frac{3}{2}$ of Fe^{5+}, respectively. $\sum\limits_{\langle i,j \rangle}$ denotes the sum over all pairs of nearest-neighbor sites i, j; each Fe^{5+} ion having six Fe^{3+} ions as nearest-neighbors, while the nearest-neighbors of

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[1]The subscripts a and f refer to antiferromagnetic and ferromagntic orderings, respectively.

each Fe^{3+} ion are three Fe^{3+} and three Fe^{5+}. The sum $\sum\limits_{\alpha,\beta}$ runs over the six different atoms in the unit cell, see figure 1.

Then to the Hamiltonian (1) we add the following non-Dirac-Heisenberg Hamiltonian of nearest-neighbor biquadratic interactions

$$H_{NDH} = -j_f \sum_{\langle i,j \rangle} \sum_{\alpha,\beta} (\mathbf{s}_{i\alpha} \cdot \mathbf{S}_{j\beta})^2 - j_a \sum_{\langle i,j \rangle} \sum_{\alpha,\beta} (\mathbf{S}_{i\alpha} \cdot \mathbf{S}_{j\beta})^2 \tag{2}$$

Note that we have adopted the convention that all exchange couplings J_a, J_f, j_a, j_f are positive.

Within the mean-field (MF) approximation it turns out that in fact the effect of adding these biquadratic interactions is to redefine the exchange integrals J_a and J_f, into temperature-dependent effective ones as follows

$$\begin{aligned}
J_a &\longrightarrow J_a^{eff} = J_a + 2j_a \langle S \rangle^2 \\
J_f &\longrightarrow J_f^{eff} = J_f + 2j_f \langle S \rangle \langle s \rangle
\end{aligned} \tag{3}$$

where $\langle S \rangle$ and $\langle s \rangle$ are the spontaneous sublattice magnetizations on the sites Fe^{3+} and Fe^{5+}, respectively. The same is also true within spin-wave theory, see sect.4. However within the linear-spin-wave approximation the effective couplings J_a^{eff}, J_f^{eff} are independent of temperature since in this case the spins S and s are respectively substituted for the sublattice magnetizations $\langle S \rangle$ and $\langle s \rangle$.

As was demonstrated by Nagaev [8], all non-Dirac-Heisenberg interactions, biquadratic in our case, change the order of the magnetic transition from second to first. It is readily seen in (3) that the effective exchange integrals decrease in magnitude with decreasing magnetizations because of a decrease in the order parameters $\langle S \rangle$ and $\langle s \rangle$. Inversely, a decrease in the effective exchange integrals in turn leads to a decrease in the magnetizations, i.e. there is a positive feedback effect. This brings about the change in the order of the transition as the ratio, $\frac{j}{J}$ in the present case, between the non-Dirac-Heisenberg and the Dirac-Heisenberg interaction reaches a certain critical value.

Henceforth, to avoid writing cumbersome formulae, we shall only give the expressions for physical quantities derived from the Hamiltonian (1), but keeping in mind that all calculations are done using the effective exchange integrals J_a^{eff}, J_f^{eff} defined in (3).

b. Spontaneous magnetizations Within the MF approximation the order parameters $\langle S \rangle$, $\langle s \rangle$ are found to satisfy the following coupled self-consistent equations

$$\begin{aligned}
\langle S \rangle &= B_S \left(z\beta \left[J_a \langle S \rangle + J_f \langle s \rangle \right] \right) \\
\langle s \rangle &= B_s \left(2z\beta J_f \langle S \rangle \right)
\end{aligned} \tag{4}$$

where $\beta = \frac{1}{k_B T}$, and $B_j(x)$ is the usual Brillouin function

$$B_j(x) = (j + \frac{1}{2}) \coth(j + \frac{1}{2})x - \frac{1}{2} \coth \frac{x}{2}$$

and $z = 3$, i.e. half the number of nearest neighbors of an atom.

Upon making the substitutions (3) the self-consistent equations (4) become more complicated and then they can be solved only numerically. The corresponding solution is shown in solid lines in figure 2, for the values of the exchange integrals obtained by fitting the experimental data on the specific heat and magnetic susceptibility, see sect.3 below.

c. Susceptibility We have computed the magnetic susceptibility both in the ordered and paramagnetic phases following the generalized mean-field approach of Smart [9]. For this purpose, we use the fact that the spontaneous magnetizations given by eqs.(4) exhibit the following antiferromagnetic-ferromagnetic arrangement at $T < T_N$ (see figure 1)

$$\langle \mathbf{S}_2 \rangle = -\langle \mathbf{S}_3 \rangle = -\langle \mathbf{S}_5 \rangle = \langle \mathbf{S}_6 \rangle, \quad \langle \mathbf{s}_1 \rangle = -\langle \mathbf{s}_4 \rangle \tag{5}$$

Thereby we obtain the following expressions for the transverse magnetic susceptibility,

$$\chi_\perp = \alpha \times \frac{J_f \left(2 \langle S \rangle + \langle s \rangle \right)^2 + 2 J_a \langle S \rangle \langle s \rangle}{2 z J_f J_a \langle S \rangle^2} \tag{6}$$

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and parallel susceptibility[2]

$$\chi_{||} = \alpha \times \frac{2T \cdot \left(2B_S'(y_0^S) + B_s'(y_0^s)\right) + 2z \cdot (J_a + 4J_f) \, B_s'(y_0^s)B_S'(y_0^S)}{T^2 + zJ_aT \cdot B_S'(y_0^S) - 2z^2J_f^2 \cdot B_s'(y_0^s)B_S'(y_0^S)} \tag{7}$$

where

$$y_0^s = (2zJ_f \langle S \rangle)\,\beta, \qquad y_0^S = z\,(J_f \langle s \rangle + J_a \langle S \rangle)\,\beta$$

and have introduced the conversion coefficient $\alpha = \mu_0(g\mu_B)^2 N_c/k_B$. With $g = 2$, and $N_c = \frac{N_A}{2}$, N_A being the Avogadro number, as there are two molecules of the sample in the unit cell.

It is worthwhile to note that the transverse susceptibility (6) is a decreasing function of temperature.

Now, as we are dealing with powder sample, the total magnetic susceptibility below the Néel temperature is given by

$$\chi(T) = \frac{1}{3}\chi_{||} + \frac{2}{3}\chi_{\perp}$$

Next, in the paramagnetic phase the total magnetic susceptibility is found to be

$$\chi_{PM} = \alpha \times \frac{2}{z} \cdot \frac{y + 2x + J_a - 4J_f}{xy + J_a x - 2J_f^2} \tag{8}$$

where

$$x = \frac{3k_B T}{z}\frac{1}{s(s+1)}, \qquad y = \frac{3k_B T}{z}\frac{1}{S(S+1)}$$

One can check that as $T \to T_N$, $B_j'(x) \to \frac{j(j+1)}{3}$, so that eq.(7) reduces to eq.(8), and at $T = T_N$, we get $\chi_{\perp} = \chi_{||} = \chi_{PM}$.

d. Entropy   Within the foregoing approach the entropy (per atom) of the system reads

$$\frac{S(T)}{N_c} = 2\left[\log\left(C_\sigma(\beta A)\right) - \beta A \langle s \rangle\right] + 4\left[\log\left(C_S(\beta B)\right) - \beta B \langle S \rangle\right] \tag{9}$$

where $A = 2zJ_f \langle S \rangle$, $B = z\left[J_a \langle S \rangle + J_f \langle \sigma \rangle\right]$, and $C_j(x) = \frac{\sinh\left[(2j+1)\frac{x}{2}\right]}{\sinh(\frac{x}{2})}$, whose derivative is the Brillouin function given earlier.

When plotted as a function of temperature, for the exchange couplings obtained below, the entropy (9) increases with increasing temperature up to the Néel point where it exhibits an abrupt jump characteristic of a first-order transition, and then it saturates to a constant given by the configurational entropy in the paramagnetic phase.

e. Specific heat   The specific heat within the same approximation is given by[3]

$$\frac{C_v}{N_c} = -2z \cdot \left(J_a \cdot \langle S \rangle \, \partial_T \langle S \rangle + J_f \cdot \left[\langle s \rangle \, \partial_T \langle S \rangle + \langle S \rangle \, \partial_T \langle s \rangle\right]\right) \tag{10}$$

for $T \lesssim T_N$.

Above T_N the mean-field approximation yields a zero specific heat since in this temperature range the order parameters and thereby the free energy vanish, see figure 4.

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[2]Here the prime stands for the derivative of the Brillouin function $B(x)$ with respect to x.

[3]∂_T stands for the derivative with respect to temperature.

III. RESULTS AND DISCUSSION

The Dirac-Heisenberg model including the biquadratic interactions studied here has been written for the ideal compound $Sr_2LaFe_3O_9$ with no oxygen vacancies. On the other hand, the compound with the closest composition to the latter was studied experimentally by Wang et al. in [4]. Therefore, we believe that our theory is more suitable for fitting the experimental data of ref. [4] than those of [2] or [3], especially from the quantitative point of view. However, we also obtain good qualitative agreement with the authors of refs. [2], [3].

A reasonable (numerical) fit to the experimental data [4] on the magnetic susceptibility and specific heat given in figures 3 and 4, led to the following values of the exchange integrals and biquadratic interactions

$$\frac{J_a}{k_B} \simeq 26K, \qquad \frac{J_f}{k_B} \simeq 6.5K \tag{11}$$
$$j_a \simeq 0.11 \times J_a, \quad j_f \simeq 0.08 \times J_f$$

As was mentioned in the introduction, the value of the antiferromagnetic exchange integral J_a found here is in agreement with the ones obtained by Anderson [10], i.e. $J_a \simeq 26K$ in $LaFeO_3$, or by Grenier et al. [11], that is $24K$ in $CaFe_2O_5$.

For these couplings we find that the sublattice magnetizations exhibit a sharp drop at the Néel temperature indicating that the transition is of first order, see figure 2. This behavior is also reflected in the magnetic susceptibility, which exhibits a sharp jump at the transition, in agreement with the susceptibility measured by Wang et al. [4] and also with the one reported by Zhou et al. [3], see figure 3. We see that the susceptibility curve obtained within the mean-field theory fits both qualitatively and quantitatively to the one measured by Wang et al. [4], however the authors of ref. [3] obtain a larger jump at the critical temperature.

In addition, the jump in the entropy (9) is indicative of a first-order transition, and yields the latent heat released by the system at the transition. Accordingly, for the exchange integrals found above, we find that the contribution of magnetic excitations to the latent heat is given by the product of the jump in the corresponding entropy at the critical temperature and the latter, i.e. $Q(T_N) = T_N \times \Delta S \simeq 2.9 \ KJ/mole$. The experimental enthalpy found by Zhou et al. [3] is $\Delta H \simeq 3.7 \ KJ/mole$, which is the total enthalpy of the system.

Consequently, the specific heat obtained within the mean-field approximation (10) diverges at the Néel temperature. In the magnetically ordered phase and around the transition we obtain a good fit of this to the experimental curve obtained by Wang et al. [4], see figure 4. As is well known the mean-field theory cannot be correct above the Néel temperature, for it predicts the absence of short-range order. In particular, in our case the specific heat computed within mean-field approximation drops to zero for $T > T_N$, since then the order parameters vanish, i.e. $\langle S \rangle = \langle s \rangle = 0$. Alternatively we have computed the contribution to the specific heat (10) in the paramagnetic phase using the approach of high-temperature expansion, but this does not yield a significant contribution. We have also taken into account that we are only dealing with the contribution of magnetic excitations to the transition while leaving out the lattice component.

To derive the Curie-Weiss law for the magnetic susceptibility in the paramagnetic phase, we note that the denominator in (8) is quadratic in temperature, and thus leads to a hyperbolic function of temperature, as in the case of a ferrimagnet. However the high temperature asymptote to the hyperbola does have the Curie-Weiss law form, that is

$$\chi = \frac{C_a}{T - \theta_a} \tag{12}$$

with

$$C_a \simeq 12, \quad \theta_a \simeq -228K$$

in fair agreement with the experimental result [2], [3]

$$C = 11.4, \qquad \theta = -250K$$

In the paramagnetic phase, we can also estimate the average magnetic moment corresponding to the average valence of iron. Indeed, the relationship between the magnetic moment per atom and susceptibility

$$\mu = \sqrt{8 \times \chi(T - \theta_a)} = 2.83 \times \sqrt{\frac{C_a}{3}}$$

yields for iron

$$\mu(Fe) = 2.83 \times \sqrt{\frac{C_a(Fe)}{3}} \simeq 5.66\mu_B$$

which agrees with the value found by Battle et al. [2], that is $5.55\mu_B$.

IV. SPIN WAVE THEORY

f. Spectrum   Within the linear spin-wave theory based on the Holstein-Primakoff representation [12] of spin operators ($\frac{1}{S}$ expansion), we find three doubly-degenerate magnon branches, as shown in figure 5. The explicit expressions of the corresponding magnon energies are rather messy and we omit writing them here. Nonetheless, around the point $\Gamma, k = (0, 0, 0)$, located at the center of the Brillouin zone we can write,

$$\hbar\omega_1(k) = \sqrt{\rho^1_{xy} \cdot (k_x^2 + k_y^2) + \rho^1_z \cdot k_z^2} \tag{13}$$

$$\hbar\omega_2(k) = \sqrt{\Delta_1 + \rho^2_{xy} \cdot (k_x^2 + k_y^2) + \rho^2_z \cdot k_z^2}$$

$$\hbar\omega_3(k) = \sqrt{\Delta_2 + \rho^3_{xy} \cdot (k_x^2 + k_y^2) - \rho^3_z \cdot k_z^2}$$

where the gaps Δ_1 and Δ_2 are given by

$$\Delta_1 = \frac{9J_f}{4} \left[2J_a \, sS + J_f(2S + s)^2 \right]$$

$$\Delta_2 = \frac{9J_f}{4} \left(2J_a sS + J_f s^2 \right)$$

and the spin stiffness coefficients ρ^1_{xy}, ρ^1_z, etc., are (cumbersome) functions of the exchange integrals. Recall that the exchange integrals J_a and J_f must be redefined using eq.(3) and taking into account the fact that in the linear-spin-wave approximation the sublattice magnetizations $\langle S \rangle$ and $\langle s \rangle$ are replaced by their nominative values $S = \frac{5}{2}$ and $s = \frac{3}{2}$, respectively.

It is seen in figure 5 that the first of the spectrum branches, the lowest curve, is gapless and of antiferromagnetic type. Whereas, the second and third branches, the upper ones, represent gapped ferromagnetic modes. We have assumed here that the anisotropy is too small to produce a gap in the magnon spetrum at temperatures the latter is obtained.

Therefore, the spectrum above shows that we have antiferromagnetic ordering at low temperature, together with a "weak ferromagnetic ordering" that starts propagating upon heating. As discussed in the introduction, it should be interesting to compare our results for the magnon spectrum with the experimental data, were Inelastic-Neutron Scattering measurements possible on the compound $Sr_2LaFe_3O_9$. This would also allow us to compare the spin stiffness coefficients we have found here with the experimental ones, and then determine the effective magnetic moments of Fe^{3+} and Fe^{5+} at zero temperature, and thereby estimate their reduction by quantum fluctuations.

g. Brillouin zone   The Brillouin zone is a $3D-$hexagon (see figure 5) whose edges are defined by

$$\frac{-\pi}{\sqrt{6}} \le k_z \le \frac{\pi}{\sqrt{6}}, \ \frac{-2\pi}{\sqrt{3}} \le k_y \le \frac{2\pi}{\sqrt{3}}, \ -\left| \frac{4\pi}{3} - \frac{|k_y|}{\sqrt{3}} \right| \le k_x \le \left| \frac{4\pi}{3} - \frac{|k_y|}{\sqrt{3}} \right|.$$

h. Thermodynamic quantities   The ground-state energy per site is given by

$$\frac{E_g}{N_c} = H_0 + \frac{1}{N_c} \frac{\sqrt{18}}{2} \sum_{\nu=1}^{3} \int_{B.Z.} \frac{d^3k}{(2\pi)^3} \, \hbar\omega_\nu(k) \tag{14}$$

where

$$H_0 = -zJ_a \cdot S(S+1) - zJ_f \cdot [S(s+1) + s(S+1)] \simeq -0.16 \ eV$$

and $\frac{E_g}{N_c} \simeq -0.12 \ eV$.

Next, within linear spin-wave theory we have studied the temperature dependence of the internal energy, specific heat, and sublattice magnetizations. We have found that at low temperatures, the internal energy behaves as T^4, and that the specific heat behaves as T^3, which is consistent with an antiferromagnetic ordering at low temperatures.

On the other hand, as discussed in the introduction, the linear spin-wave theory turns out to be a poor approximation in the vicinity of the transition, as long as sublattice magnetizations are concerned. Indeed we find that $\langle S \rangle$ and $\langle s \rangle$ decrease linearly with temperature as this approaches the Néel point, and vanish at different temperatures, see figure 2. There we also see that the linear-spin-wave theory yields, at zero temperature, quantum corrections to the magnetic moments of Fe^{3+} ions but no corrections to those of Fe^{5+} ions. Therefore, we think that further non-linear corrections should be taken into account so as to obtain the correct temperature dependence of the sublattice magnetizations especially as the temperature approaches the Néel point.

V. CONCLUSION

We have modelled the first-order magnetic transition occurring in the perovskite $Sr_2LaFe_3O_9$ using a Dirac-Heisenberg Hamiltonian including (nearest-neighbor) biquadratic interactions, whose origin could be related with the disproportionation of iron. We have been able, by fitting the experimental magnetic susceptibility and specific heat, to estimate the superexchange integrals for the antiferromagnetic and ferromagnetic interactions, as well as the biquadratic ones. The first of these integrals is consistent with the results of previous work on kindred compounds.

Elastic-Neutron Scattering will be performed on this compound for checking the anomalous behavior of the sublattice magnetizations predicted here. Unfortunately, it is difficult to check up on the magnon spectrum we have computed. Nevertheless, the latter does confirm the magnetic structure determined from Neutron-Diffraction data by Battle et al. [2] in the compound $Sr_2LaFe_3O_{8.94}$.

Non-linear spin-wave calculations and Monte Carlo simulations are still under investigation. Finally, it should be very instructive to understand more in detail the effect of iron disproportionation, or more generally the charge redistribution, on the magnetic ordering.

VI. ACKNOWLEDGMENTS

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Figure captions

- Figure 1: The crystal and magnetic structure of $Sr_2LaFe_3O_9$ [2]. The large circles represent Fe^{3+} and the medium ones represent Fe^{5+}, with the arrows indicating the size and orientation of their spins; the smallest circles stand for intermediate oxygen atoms. The $2Sr^{2+} : La^{3+}$ cations which are disorderly located in the cubic centers have been omitted for clarity. The hexagonal unit cells of the two structures are commensurate, but the unit cell of the crystal structure is a triple one, while that of the magnetic structure is a primitive one, for the rhombohedral translations are absent in the magnetic structure. Each unit cell contains two formulae of $Sr_2LaFe_3O_9$, hence six iron ions, which form six magnetic sublattices below the ordering temperature.

- Figure 2: The solid curves represent the temperature dependence of the sublattice magnetizations predicted by mean-field theory including biquadratic interactions. The upper (solid) curve represents the magnetization $\langle S \rangle$ of the Fe^{3+} ions of spin $\frac{5}{2}$, and the lower curve is the magnetization $\langle s \rangle$ of Fe^{5+} of spin $\frac{3}{2}$, obtained for the exchange integrals given in eq.(11) in the text.

  The dashed curves represent the corresponding sublattice magnetizations predicted by the linear-spin-wave theory, up to a temperature of $120K$.

- Figure 3: The curve in balls represents the experimental magnetic susceptibility of ref. [4], and the one in triangles is the suscpetibility measured by Zhou et al. [3]. The solid line is the magnetic susceptibility obtained from MF theory including biquadratic interactions.

- Figure 4: The curve in balls is the experimental specific heat of ref. [4], and the solid line is our theoretical result of MF theory including biquadratic interactions.

- Figure 5: Plot of the magnon spectrum along the path $Z\Gamma XM\Gamma Y$ indicated in the Brillouin zone shown as inset.